BETA DECAY OF NEUTRON-RICH TRANSURANIC NUCLEI

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Beta Decay of Neutron-Rich Transuranic Nuclei*

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The intense neutron flux generated in a thermonuclear explosion is of interest to nuclear physicists because, among other reasons, the atoms exposed to this flux will undergo many successive capture reactions during the brief period the flux exists. If 238U target atoms are given such exposure, the phenomenon becomes a way to probe the nuclear properties of very neutron-rich uranium nuclei and to produce heavy isotopes of the transuranium elements that survive the chains of beta decay following the initial phase of the reaction. This unique method of transuranium element production was investigated by U.S. scientists in a series of thermonuclear explosions during the period 1952-1969, first as an unexpected result of the Mike explosion [1] that occurred in November 1952 and later in a series of underground explosions where the explosive devices were designed specifically for this purpose [2,3]. The heaviest species identified in this work was 257Fm, which implies the production of 257U during the capture phase. While nuclei this neutron-rich cannot be produced by any other man-made technique, it is known, of course, that such nuclei are generated by the astrophysical r process. The neutron-capture path for this process is thought to pass through a region around 262U[4]. Given the clear relationship between these processes, it is the purpose of this paper to see what insights into recent r-process calculations can be gained from a consideration of the experimental data for heavy element production in thermonuclear explosions.

Experimentally determined mass-yield curves for several underground thermonuclear explosions are shown in Fig. 1. The curves are all similar in form, each with the following characteristics: 1) A regular, exponential decrease with increasing A; 2) Data points often missing for either A=249 or A=251, which is due to the circumstances of the experimental measurements; 3) No data shown for A < 244 (or perhaps 242). Although such nuclides can be readily determined, it was often impossible to deduce the fraction of observed atoms arising from multiple neutron capture processes in a 238U target due to interference from large quantities of residual plutonium isotopes; 4) Termination of the yield curve data at A=257; 5) Some odd-even variation within the curve. For all of the data in the mass range A=244-248, the observed yields of the even-A products are systematically higher than those for the odd-A products. Beginning somewhere near A=250, this odd-even variation is seen to be reversed at higher masses. The only exception to this observation is the Hutch mass yield curve where no reversal is observed up to A=255.

As a starting point for calculating product yields, we will consider a simple model in which the assumed capture sequence begins with a single target isotope and consists of isotopes of the same element, all with identical capture cross sections(σ). Losses due to processes other than capture (e.g. neutron-induced fission) are neglected. For such a system the yield of the nth member in the sequence is given by the following expression:

$$N_n = N_0(0) \exp(-\sigma\Phi) (\sigma\Phi)^n / n! \qquad (1)$$

where $N_0(0)$ is the amount of target material at zero time,

and   Φ is the neutron exposure, i.e. the product of
neutron flux (n/cm^2) and time(t).

This expression describes a family of smooth mass-yield curves, all concave downwards and whose slopes at any given mass number tend to decrease with increasing values of neutron exposure.
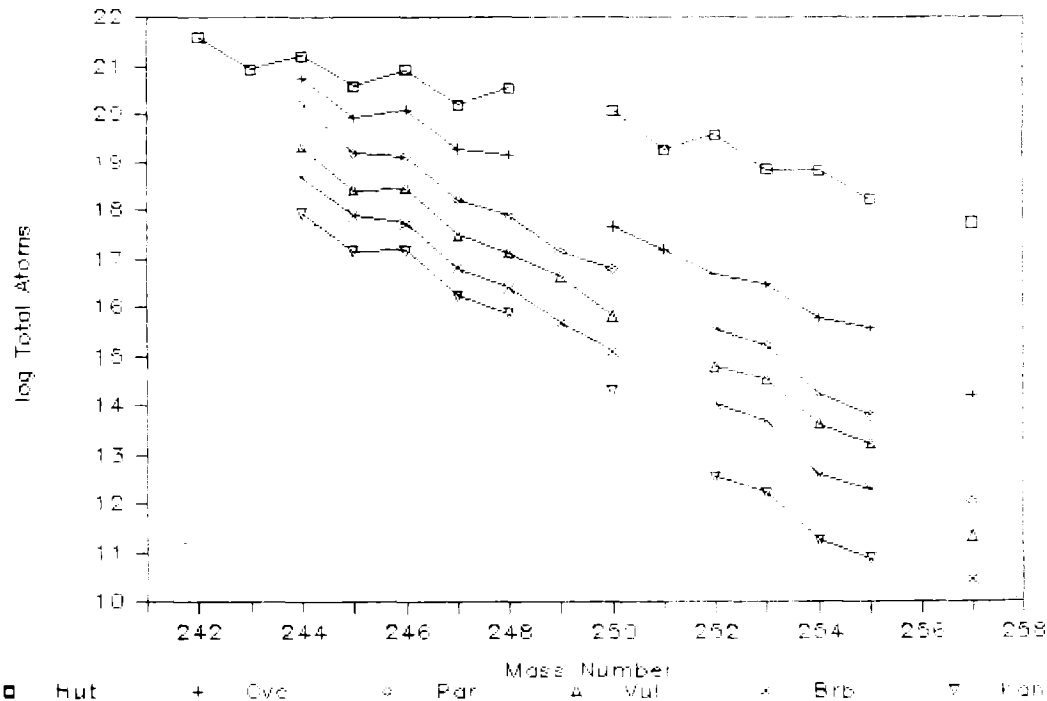

Fig.1 Mass-yield curves for several underground explosions. Except for Hutch, the relative positions of the curves on the ordinate have been adjusted to facilitate comparison; the true position of each curve is given by the absolute amounts of mass 244 atoms produced, in units of 10^{20} atoms: Hutch 17, Cyclamen 3.8, Par 8.7, Vulcan 2.7, Barbel 0.10, and Kankakee 11.

By departing from these simple assumptions, the odd-even variation within a given curve is readily explained by a corresponding variation in capture cross section for odd-mass and even-mass isotopes. This is exactly the expected behavior for the capture cross sections because of an odd-even alternation in neutron separation energies, a natural manifestation of the nucleon pairing phenomenon. On the other hand, understanding the linear nature of the observed yield curves presents a somewhat greater challenge. It was this feature of the mass-yield curve from the Mike event that led CAMERON [5] to suggest for the mass equation used for predictions in this neutron-rich region a revised form which provided for a less rapid decrease in the neutron separation energy with increasing neutron number. Nevertheless, since it is known that the overall trend for separation energies is one of decrease and that eventually neutrons are no longer bound (at the neutron drip line), it is not possible, within the simple model considerations discussed here, to produce a calculated mass-yield curve that is linear over an extended range of masses. For this reason, the observed mass yield curves in Fig. 1 were thought to be the envelope of several separate curves.

This idea has been invoked, in fact, to explain the existence of the reversal in the odd-even alternation observed in the majority of the data (Fig.1). It was proposed that the nuclides observed beyond A=250 were produced by sequential neutron capture in an odd-Z isotopes. The odd-even effect is reversed because now capture cross sections for even-mass isotopes (which are odd-odd species) will be larger than for odd-mass isotopes. BELL [6] has shown that an initial production

of 238Pa and/or 239Np can occur by (n,p) and (d,n) reactions on the 238U target before moderation of the source neutrons. Although these odd-Z nuclides would be present in much lower abundance (approx. 0.01) than the 238U, the larger cross sections of the capture sequence eventually cause the yields from the odd-Z chain to surpass those from the uranium chain. INGLEY [3] has made quantitative fits to most of the data of Fig. 1, deriving best values for effective capture cross sections (at 20 keV), neutron exposure values for each event, and ratios of initial amounts of 238Pa and 238U required to reproduce the mass-yield curves.

Another variable studied in the underground explosion series was that of the composition of the initial target material. It was found that the greatest success was obtained with 238U, although various other heavy nuclides such as 243Am, 242Pu, 237Np, and 232Th were incorporated in the targets of various test devices [2,3]. Neutron-induced fission presumably was the cause of low production for some of the heavier alternate target materials.

A disappointing feature of the experimental study of these products from thermonuclear explosions was the inability to detect evidence for nuclides with A>257. Certainly the trends of yields for the higher exposures (e.g. in Hutch and Cyclamen) were such that one would predict production of detectable amounts of species up to A=260 or even higher, depending upon specific decay properties. This absence of heavier isotopes can be explained by at least three loss mechanisms: 1) Neutron-induced fission competing with capture, 2) Beta-delayed fission in the decay chain, and 3) Short half-lives of the isotopes at the end of the decay chain. This last mechanism apparently can account for the inability to detect species with A=256, 258, and 259. The beta-stable nuclides at the end of these decay chains are now known to have very short spontaneous fission (SF) half lives [7-9], viz. 12-m 256Cf, 0.4-ms 258Fm, and 1.5-s 259Fm.

Of greater interest to this question has been the recent discovery [10] of a 32-day SF activity assigned to 260Md. Although it appears that the experimental techniques used in the 1960's would have permitted detection of 260Md, had it been present in the samples then, there are two arguments suggesting it was not present: 1) The end of the mass 260 beta minus decay chain is calculated to be 260Fm; 2) Even if 260Fm proved to be unstable towards beta minus decay, it is unlikely that the recently discovered 260Md activity would have been produced. The argument here is that the relatively long half life of the observed 260Md, coupled with a decay energy of 0.5-1.0 MeV, suggests considerable hindrance of its beta decay (log ft >8.5), probably due to its high spin. If this is so, the 32-day activity will not be populated at the end of a beta-decay chain because the successive decays, which are populating even-even and odd-odd nuclides alternately, will immediately revert to populating only low-spin levels once the decay has passed through the first 0^+ even-even ground state in the chain.

In the past 10-15 years, advances in the formulation of beta-strength functions and in the extension of mass formulae and fission barrier calculations away from beta stability have resulted in several papers that treat the phenomenon of beta-delayed fission (BDF) and beta-delayed neutron emission in neutron-rich nuclides. With the availability of quantitative estimates of these phenomena, allowance has been made for their effect on the calculated abundances of nuclides in beta-decay chains in the astrophysical r-process, and, less frequently, on the observed abundances of products from thermonuclear explosions. We will consider here the effect of BDF in this latter application by assessing the results of four separate calculations, the elements of which are listed in Table 1. Beta-delayed neutron emission will not be considered, at least partially because its effect is to shift atoms from one beta-decay chain to the next and, thus, to some extent to nullify any loss by replacement of atoms from the next higher chain. We have calculated correction factors for each mass chain by taking the product of the survival fraction for each decay step in the chain. These correction factors, which are shown in Fig. 2, were then applied to the Hutch mass yield curve with the results shown in Fig. 3.

Table 1. Calculations of beta-delayed fission and r-process abundances

| Beta-Strength Function	Mass Formula	Fission Barrier
WENE and JOHANSSON [11] Constant, i.e. prop. to daughter level density	JOHANSSON and WENE [12]	Statistical, employing fission, neutron,and gamma widths
KODAMA and TAKAHASHI [13] Gross theory of beta decay (no level density formula required)	MYERS and SWIATECKI [14]	Systematic formulation based on exp. data
THIELEMANN [4] Tamm-Dancoff approx. with long-range GT residual interaction	HILF [15], (Fig. 5a [4]) VON GROOTE [17] HOWARD and MOELLER [16], (Fig. 5c [4])	HOWARD and MOELLER [16]
MEYER [18] Nilsson RPA treatment with infinite range GT interaction [19]	HOWARD and MOELLER [16]	HOWARD and MOELLER [16]
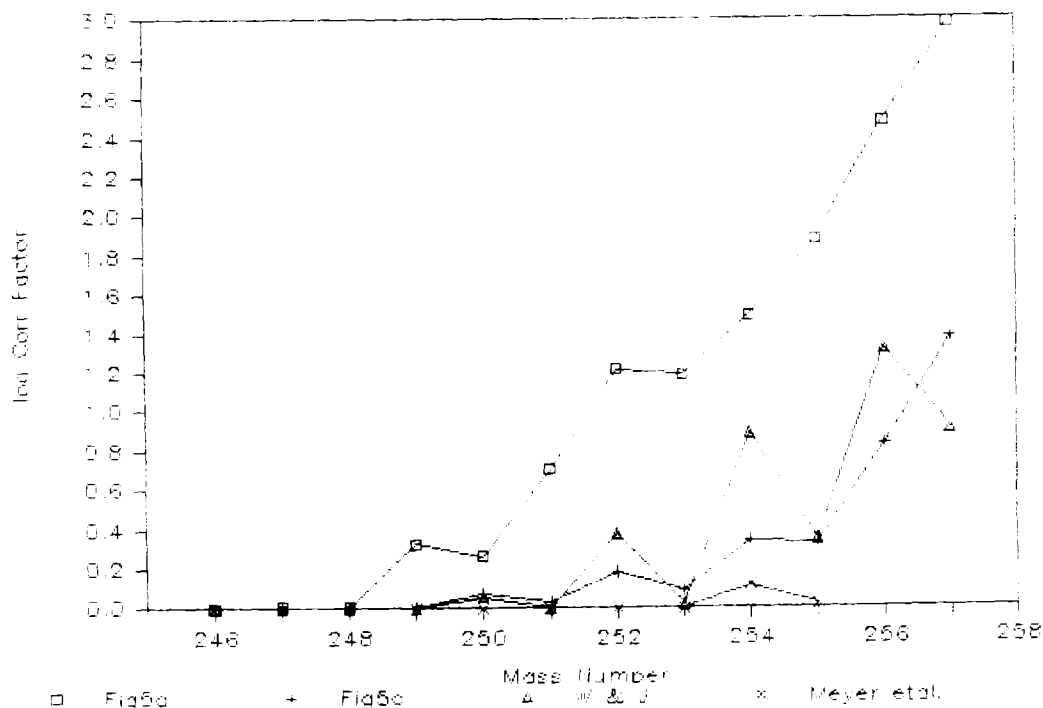
Fig.2 Beta-delayed fission correction factors for decay chains beginning with uranium isotopes.

The largest amount of BDF is calculated by THIELEMANN [4], hereafter referred to as TMK, using the mass formula of HILF [15]. When these BDF survival factors are applied as corrections to the Hutch mass-yield curve, the new data set (labelled as "Fig.5a" in the legend of Fig. 3, referring to a figure in TMK) indicates substantially greater yields than before for nuclides with A>250. The correction to the A=257 point is so great that its yield is second only to that
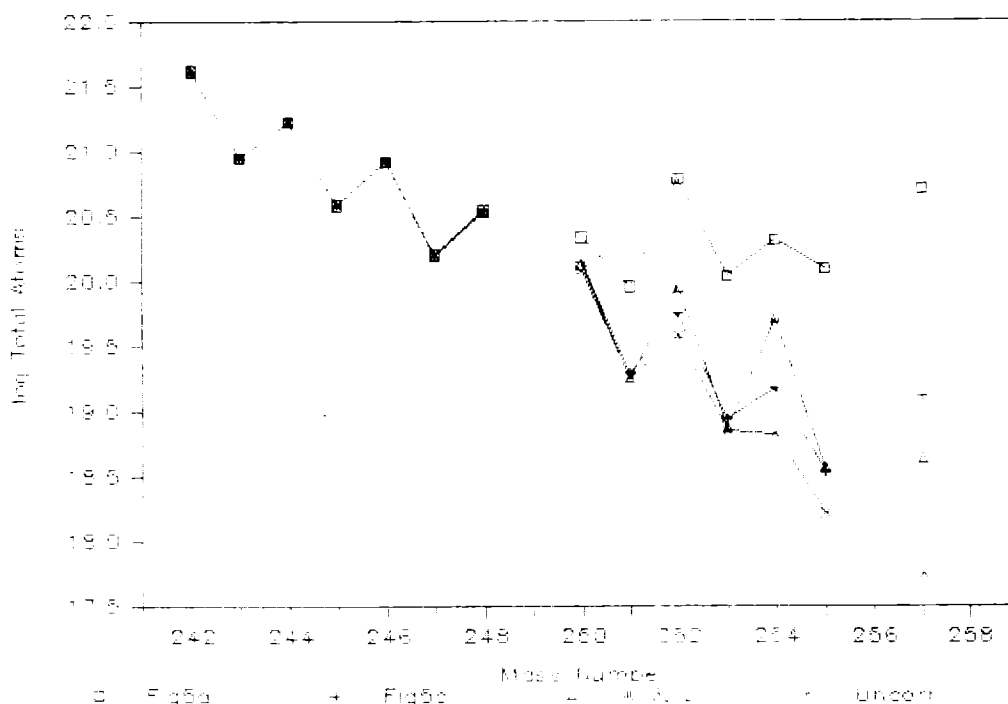
Fig.3 Hutch mass-yield curve corrected for beta-delayed fission.

for A=252 for all masses with A>246. The capture cross-section values required to produce a calculated fit to these data will be much larger for A>250 isotopes than deduced previously. In fact, we expect these revisions to require cross-section values so large as to be unphysical. There is nothing in the trend of calculated neutron binding energies that would suggest enough perturbation to permit large increases in the calculated cross sections.

A second set of results from TMK (labelled in the legend as "Fig.5c") is also shown in Fig. 2; these can be compared with the calculations of MEYER [18] where common elements are the mass formulae and fission barrier calculations. One can see the correction factors of MEYER [18] are negligible on the scale of Fig.2, except for A=254, while those of TMK for A=252-255 are as large as a factor of 2 and rise rapidly to even greater values for A=256, 257. The differences here, which are a function only of the assumed form of the beta-strength function, show that TMK's treatment tends to place a greater fraction of the beta strength in excited levels near or above the fission barrier than does that of MEYER [18].

Those BDF calculations that produce large even-odd variation, e.g. see the data labelled "W&J" in Fig. 2 (from Wene and Johansson [11]), can account for the reversal of this variation in the mass-yield curves of Fig. 1. However, large even-odd variations are not a feature of all BDF calculations. In all cases, the TMK calculations show that the correction factors become extremely large (>1000) for decay chains with A>or=258; in fact, their calculations indicate the beginning of a region at A=258 where most nuclei will exhibit 100% BDF, and atoms entering this region will not survive. On this basis, they predict no production of superheavy elements by the r process. KODAMA and TAKAHASHI [13] found that BDF was negligible for Z<94, but was appreciable in heavier nuclei. For the only examples given in the paper, they indicate little significant BDF for neutron-rich species with A=266-270, excepting 269Lr and 270No, for which BDF branching was 35-40%.

Thus, the four papers cited in Table 1 include calculated estimates of beta-delayed fission with widely varying magnitudes, an indication of the degree of uncertainty inherent in extrapolating the various basic parameters. Using the effect of BDF corrections on the mass-yield curves considered in this paper as a general indication of their accuracy, we reach the following conclusions: 1) The TMK treatment with the Hilf mass formula definitely overestimates BDF for uranium decay chains in the A=244-257 region. Paradoxically, TMK considered the Hilf mass formula most realistic for neutron-rich nuclides and used these BDF survival factors to modify their r-process calculations. Some of the other theoretical estimates summarized in Table 1 may also overestimate BDF; 2) The effect of BDF can be invoked to explain two features of the thermonuclear explosion mass-yield curves, the reversal of the odd-even yield variation around A=250 and the termination of the curves at A=257. On the other hand, since there are alternate possibilities that explain the observations adequately, the presence of substantial amounts of BDF is not required in order to understand the data.

Since the results of BDF calculations show much variability and, thus, there is no indication that they are accurate, it seems wise to assess their effect on r-process calculations rather carefully. Therefore, it would be useful to determine the sensitivity of calculations of r-process abundances and r-process production ratios for chronometric pairs to the magnitudes and structure of various calculations of the BDF phenomenon.

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